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16. ABSTRACT The potential health problems associated with the presence of persistent organic chemicals in wastewater and sludge, when applied to agricultural lands, are reviewed. The topics considered include: the type and amounts of organic chemicals present in wastewater and sludge, their fate on land, potential health effects on workers and other exposed populations, and effects on the quality of ground and surface waters and on the food chain. Because information on the effects of chronic exposures is scarce, several examples are given from cases of acute exposure, usually to wastewater treatment plant workers. There is insufficient information available to assess the longterm health risks of exposure to organics in wastewater treatment plants or at land application sites. Further research is recommended on the uptake of organic chemicals by food crops. Long-term follow-up is recommended for populations who have had acute short-term exposure to organic chemicals from waste materials.

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HEALTH RISKS OF ORGANICS IN LAND APPLICATION

By Vimala A. Majeti[1] and C. Scott Clark[2]

INTRODUCTION

Among all constituents of concern in wastewaters and sludges, the effects of the toxic organic chemicals are the least known. Most attention has thus far been paid to the fate and effect of heavy metals, but little to the impact of persistent organic compounds. Since many heavy metals and organic chemicals will be in more concentrated form in sludge than in wastewater, the latter may be generally considered less hazardous than sludge, but there may be exceptions. Topics considered in this review of persistent organic chemicals are: (1) Potential problems to workers and other populations with wastewater/sludge contact during wastewater treatment, transportation, or application; (2) effects on the food chain, and on the quality of ground and surface waters; and (3) recommendations for regulation. Little information is available on direct health effects of organic chemicals in wastewater and sludge. Therefore, examples of health effects of wastewater treatment plant workers from acute exposure to toxic organic chemicals are included where applicable, to provide some information regarding possible consequences of worst case atmospheric exposure at land application sites.

In order to assess the potential health risks of the organic chemicals during the practice of the various forms of land application and disposal, a number of factors need to be understood:

1. The type and amounts of the organic compounds present in wastewaters and sludge.

[1]Research Assoc., Dept. of Environmental Health, Univ. of Cincinnati, 3323 Eden Avenue, Cincinnati, Ohio 45267.
[2]Assoc. Prof., Dept. of Environmental Health, Univ. of Cincinnati, 3323 Eden Avenue, Cincinnati, Ohio 45267.

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2. Their fate and potential environmental contamination, as a result of application processes.
3. Available control measures.
4. Their effects on wastewater treatment plant workers, on populations living near land application facilities, on ground and surface water pollution, and on the food chain.

Characterization of Organic Chemicals in Wastewater and Sludge

The presence of organic matter in raw and treated wastewater is highly variable and dependent upon the source of wastewater and the degree of treatment. Attempts to characterize organic matter in wastewaters in the past have not been very successful. It is only recently that the extensive identification of organic compounds in waters, drinking water in particular, has been undertaken. A summary listing of some 1000 organic compounds in effluent waters has been published by World Health Organization (WHO) (69). Many of these organics have been identified in domestic sewage. Some of these compounds are very toxic, and some of them are known or suspected carcinogens as reviewed in a research report to Congress by the National Academy of Sciences, National Research Council (42). Chlorination of treated wastewater results in the formation of a wide range of chlorinated derivatives (37). Many of the organic chemicals present in wastewater are toxic and potentially carcinogenic, teratogenic, or mutagenic to humans. Relatively large numbers of organic compounds survive the conventional treatment process and are present in the treated effluent in the μg/L range. The sources of these compounds are difficult to trace except when there is an accidental spill, such as from local industrial plants.

In the past few years there have been significant efforts toward determining the nature and concentrations of the organic compounds present in wastewaters and sludges. Some work in this area has been devoted to chlorinated hydrocarbons, pesticides, and PCBs. The United States EPA Office of Research and Development has been conducting a survey of chlorinated hydrocarbons in municipal wastewater effluents which showed the presence of chlorophenols, chlorobenzenes, chloroalkanes, and chloroalkenes (54). Table 1 lists some of the organic compounds together with the range of concentrations found in municipal wastewater effluents (25,55,56). The concentrations of chlorinated hydrocarbon pesticides and other organic compounds identified in several wastewater sludges are presented in Table 2 (25,33).

Municipal sludges from Schenectady, N.Y., and Bloomington, Ind. were found to contain 23.1 ppm (18) and 300 ppm (26) PCBs, respectively. General Electric Company in Schenectady, New York, and Westinghouse Electric in Bloomington, Ind., both manufacture electrical capacitors in which PCBs are used as insulating fluids. Municipal sludge in Hopewell, Va., was found to be contaminated by kepone, a chlorinated pesticide manufactured by Life Science Products which discharged kepone waste into the city sewage system (27). Kepone levels of 0.1 ppm to 10 ppm were found in the sludge samples from the holding pond and from the landfill near the Hopewell Sewage Treatment plant. The concentration of two PCBs in sludge from a number of municipal wastewater plants in Michigan ranged from less than 0.1 mg/L to 352 mg/L (16,34). As would

be expected, the concentrations of the organics are considerably greater in sludge than in wastewater.

In early 1979 in Adrian, Mich., the wastewater and sludge in the wastewater treatment plant, Raisin River water and sediment, roadside soil, garden soil, and air were found to be widely contaminated with Curene 442 (MOCA, 4,4′-Methylene-bis (2-chloroaniline), a potential human carcinogen (35). Curene 442 is used as a curing agent for isocyanate-containing polymers and polyurethane

TABLE 1.—Ranges of Concentrations of Organic Compounds in Municipal Wastewater Treatment Plant Effluents (25,55,56),[a] in micrograms per liter

Compound (1)	Dayton, Ohio[b] Unchlorinated (2)	Dayton, Ohio[b] Chlorinated (3)	Cincinnati, Ohio[c] Unchlorinated (4)	Cincinnati, Ohio[c] Chlorinated (5)	Muskegon, Michigan[d] (6)
Chloroform	0.3 to 1.4	0.4 to 12	0.1 to 0.7	0.5 to 12	1 to 13
Trichloroethylene	0.2 to 1.7	0.1 to 10	—	0.6 to 0.7	2 to 10
Benzidine	—	<0.1	—	<0.1	<1
Vinyl Chloride	—	<1	—	<1	<1
Benzene	—	0.2 to 40	—	0.3 to 3.8	<1 to 8
PCBs	—	<1	—	—	<10
Endrin	—	<1	—	—	<1
Toxaphene	—	<1	—	—	<50
Methanol	—	150 to 510	—	•	—
Ethanol	—	150 to 3,000	—	•	—
Acetone	—	50 to 300	—	20 to 400	—
2,3-Dithiabutane	—	•	—	1	—
Carbon Disulfide	—	•	—	2 to 8	—
1,1,1-Trichloroethane	—	1 to 15	—	•	—
Tetrachloroethylene	—	1 to 20	—	0.3 to 3	—
Toluene	—	1 to 10	—	1	—
Xylene	—	1 to 15	—	10	—
Acrolein	—	20 to 200	—	10 to 150	—
Acetaldehyde	—	90 to 1,350	—	100 to 560	—
Carbon Tetrachloride	—	3	—	•	—
Chlorodibromomethane	•	0.1	0.1	0.4 to 4.6	—
Dichlorobromomethane	0.1	0.1 to 0.4	0.1 to 0.3	0.1 to 8	—
Bromoform	•	0.1 to 0.3	•	0.2 to 0.3	—
1,3-Dichloroethane	1.4	0.1 to 4.6	•	•	—
Methylene Chloride	—	2 to 50	—	1 to 10	—

[a] Adapted from "Risk Assessment and Health Effects of Land Application of Municipal Wastewater and Sludges" with permission of B. P. Sagik and C. A. Sorber, eds., The University of Texas at San Antonio, San Antonio, Tex. (1978).

[b] From EPA (55). Range of values for samples having concentrations above the detection limit in chlorinated and unchlorinated effluent from Dayton secondary (trickling filter) sewage treatment plant, mixed domestic and industrial wastewaters.

[c] From EPA (55). Range of values for samples having concentrations in chlorinated and unchlorinated effluent from Cincinnati's Muddy Creek secondary (AS) sewage treatment plant; primarily domestic wastewater.

[d] From EPA, Robert S. Kerr Research Laboratory (56). Range of concentrations in five samples of effluent from subsurface drainage tiles under the corn fields at the spray irrigation wastewater management system in Muskegon, collected on five days in fall 1976 (minimum detection limit approx 1 μg/L).

Note: • indicates none detected; dash (—) indicates no data available.

TABLE 2.—Concentrations of Organic Compounds in Domestic Sludges, in micrograms per liter of Liquid Sludge (9,25,33)[a]

Compound (1)	North Toronto[b] (2)	Point Edward[b] (3)	New-market[b] (4)	Sarnia[b] (5)	MSDGC[c] (6)
Solids in sludge, as a percentage	6.25	6.90	12.2	16.19	
Chlorinated hydrocarbon pesticides and PCBs:					
pp′ DDT	1.33	0.77	1.12	2.70	—
op′ DDT	6.87	4.11	1.15	5.25	—
pp′ DDE	7.18	3.68	2.18	20.77	—
pp′ DDD	4.31	3.98	1.60	6.41	—
Total DDD/DDE/DDT	19.69	12.54	6.68	35.13	—
DDT	—	—	—	—	<100
a-Chlordane	15.79	12.51	5.69	16.99	—
γ-Chlordane	12.56	11.41	2.84	12.92	—
Total chlordane isomers	28.35	23.92	8.53	29.91	<10
Heptachlor	5.37	2.29	0.70	14.95	<200
Heptachlor epoxide	3.04	1.77	0.58	6.78	<200
Total heptachlor	8.41	4.06	1.28	21.73	—
Aldrin	2.03	1.09	0.70	9.40	<10
Dieldrin	3.03	1.69	1.65	3.23	<10
Lindane	1.48	1.16	0.63	3.22	<500
Toxaphene	—	—	—	—	<10
Total organochlorine pesticides	63	45	20	103	—
PCBs (Arochlor 1254)[d]	214	108	74	1,122	—
Other pesticides and organics:					
Orthophosphates and carbamate insecticides	—	—	—	—	<10
2,4-D	—	—	—	—	<1,000
2,4,5-T	—	—	—	—	<5
2,4,5-TP	—	—	—	—	<30

[a]Reprinted from "Risk Assessment and Health Effects of Land Application of Municipal Wastewater and Sludges" with permission of B. P. Sagik and C. A. Sorber, eds., The University of Texas at San Antonio, San Antonio, Tex. (1978).

[b]Chawla, et al. (9). Mean levels of organochlorine pesticides in digested chemical sludges in four Ontario treatment plants using lime (Newmarket), iron (North Toronto, Sarnia) or alum (Point Edward) treatment.

[c]McCalla, et al. (33). Concentrations in sludges from five sludge sources in the Metropolitan Sanitary District of Greater Chicago (MSDGC), solid content of the sludges unknown.

[d]Mean monthly levels (33).

Note: Dash (—) indicates none detected.

elastomers. The source of contamination at the Adrian Wastewater Treatment Plant was traced to discharges received from the Anderson Development Company over a period of several years. Curene 442 is not very soluble in water and therefore tends to accumulate in sludges and soils. Anderson Development Company discontinued production of Curene 442 in February, 1979 by order of Michigan Department of Natural Resources.

Although the list of compounds shown in Tables 1 and 2 is quite extensive it is by no means exclusive, as to either the total number or types of organic compounds present. At present the identification of organic compounds in wastewaters and sludges is limited by the volatility of the compounds and the available analytical techniques. Most organic compounds which come directly from industrial effluents or are metabolites or degradation products cannot be readily detected or determined by the available analytical procedures. This is especially true for compounds with high molecular weights and for more hydrophylic compounds.

Fate of Organic Chemicals on Land

Several investigations have been conducted to determine the fate of disease-causing microorganisms, nitrogen, and phosphorus in the wastewater during the course of land application (8,67). There are few studies at the present time about the fate of potentially toxic chemicals in the wastewater and their possible pathways in soils. Hazardous substances in wastewater that are not removed during the treatment process may pose a present or potential long-term danger to human health or to other living organisms because they are often nonbiodegradable, bio-accumulate, or persistent in the environment.

The concentrations of organic chemicals are usually low (in micrograms per liter) in treated wastewater effluents and sludges. Repeated application on land may result in accumulation of organic chemicals in soil. The ionizability, water solubility, and structure-function relationships (66) of the organic chemicals determine to a great extent their ultimate fate and behavior in soil and the environment.

The behavior and decomposition pathways of many pesticide residues which constitute a significant fraction of the persistent organics in soils have been studied for many years (21). Organic chemicals in wastewater and sludge applied to land could be expected to be influenced by the same processes that affect the fate and behavior of organic pesticides. Some of the processes that can cause the breakdown of pesticides are photodecomposition, chemical and microbiological decomposition, and detoxification by crop plants or weeds. Several factors such as pH, surface activity, and solubility affect the degradation rate. Microbial degradation is influenced by the microbial population and by factors affecting microbial activity. Simple organic compounds such as aliphatic and phenolic acids, amino acids, and sugars are readily decomposed by soil microorganisms. Highly chlorinated or halogenated compounds, and compounds with branched chains and higher molecular weight, are more resistant to biodegradation and require longer periods of time to decompose in soils. Transfer processes move and dilute the pesticides, keeping their chemical structures intact. These processes include: (1) Volatilization; (2) movement into and out of plants by adsorption and exudation; (3) retention by crop plants and weeds in small amounts;

(4) runoff into streams and lakes; (5) movement downward into the soil in percolating water and upward from lower depths by capillary flow; and (6) adsorption and inactivation by soil constituents (67).

The adsorption phenomena of soils could increase the organic chemical concentration at the soil surface and, therefore, make them more susceptible to microbial decomposition. Both laboratory and field studies indicate that many pesticide residues adsorb to surface soils with only a few migrating to depths of 300 mm to 600 mm in soil (2). Besides adsorption of organic pesticides and other chemicals by soil, chemical reaction with functional groups on humates can also occur. The immobilized organic substances in soils are subsequently subject to chemical or photochemical decomposition, volatilization, and microbial decomposition.

The pesticides endrin, lindane, methoxychlor, toxaphene, 2,4-dichlorophenoxy acetic acid (2,4-D), and 2-(2,4,5-trichlorophenoxy) propionic acid (2,4,5-TP, Silvex) were analyzed in soil samples at a long-term land application site in Vineland, N.J., by Koerner and Haws (28). Lindane and 2,4,5-TP were found to be present at higher concentrations at depths to 310 mm at the land application site than at control sites ($p = 0.05$).

TABLE 3.—Aeration Basin Influent and Effluent, and Air Concentrations of Selected Chlorinated Organics Near Aeration Basins (11,22)

Sample (1)	Chloroform (2)	Tetrachloro-ethylene (3)	Trichloro-ethylene (4)	Trichloro-ethane (5)
Aeration basin influent[a]	480 to 1,520	<0.1 to 92.5	<0.1 to 118	<0.1 to 72
Aeration basin effluent[a]	18 to 82	<0.1 to 88	<0.1 to 140	<0.1 to 17
Air samples downwind of aeration basins[b]	7 to 202	8 to 46	3 to 73	10 to 90
Threshold Limit Value (TLV)[b]	50,000	670,000	535,000	1,900,000

[a]Quantities of compounds given in micrograms per liter.
[b]Quantities of compounds given in micrograms per cubic meter.

A study conducted at Muskegon, Mich., by the United States Environmental Protection Agency focused on the behavior of a number of organic compounds in the land application system (56). The Muskegon County Wastewater Management System is an aeration, lagoon impoundment, and spray irrigation facility (14). The study consisted of analysis of raw influent, aeration effluent, storage lagoon effluent, and irrigation field drain tile effluent for several organic compounds including benzene, chloroform, trichloroethylene, vinyl chloride, benzidine, endrin, toxaphene, and PCBs. A total of 64 organic compounds were identified in the influent and various effluent samples (the minimum detectable limit is approximately 1 μg/L). Some compounds including toluene, 1,2-dichloroethane, xylene, diazobenzene, dichlorobenzophenone, chloroaniline, hexadecanoic acid, octadecanoic acid, dodecanol, and tetradecanol are believed to be present in relatively higher concentrations, possibly several hundred micrograms per liter or more, in the influent wastewater. The study showed that benzene, chloroform, and trichloroethylene were present in influent raw

wastewater in concentration ranges from 6 μg/L to 53 μg/L, 360 μg/L to 2645 μg/L, and 6 μg/L to 120 μg/L, respectively. Concentrations of these compounds were significantly reduced in the treatment sequence. Low levels of only a few of the total identified 64 compounds were detected in the final effluent from the system. The Muskegon system appears to be relatively effective in removing organic pollutants from the wastewater. Although removed from the raw influent these compounds are mostly concentrated in the lagoon sediment.

Another study conducted at Muskegon, Mich., involved monitoring worker exposure to several volatile organic compounds, bacteria, and viruses in aerosols (11,22). The compounds analyzed for in the air and wastewater samples included chloroform, carbon tetrachloride, tetrachloroethylene, trichloroethylene, and trichloroethane. Carbon tetrachloride was not detected in any of the samples collected. The concentrations of the other four compounds in aeration basin influent and effluent, and in air samples collected downwind of the aeration basins, are shown in Table 3. As can be seen from Table 3, the aeration process reduced the wastewater concentrations of chloroform and trichloroethane, but did not have much effect on the concentration of tetachloroethylene or trichloroethylene. The downwind air concentrations of these compounds indicate that significant quantities are volatilized during the aeration of wastewater. It must be emphasized that for all four compounds, the levels in air are far below the established threshold limit values (TLVs) (Table 3) (52). Analysis of air samples around the spray irrigation field rigs showed relatively low levels of tetrachloroethylene, trichloroethylene, and trichloroethane, but no chloroform. This is believed to be due to the relatively low concentrations of these compounds in the water being applied.

Effect on Wastewater Treatment Plant Workers

The workers at the wastewater treatment plant in Adrian, Mich. were exposed to Curene 442 [MOCA, Methylene-bis(2-chloroaniline)] in wastewater and sludge over a period of several years (35). The source of Curene 442 is discharges from Anderson Development Company in Adrian, Mich. Based on tissue culture carcinogenicity tests and animal test studies in mice, rats, and female beagle dogs, National Institute of Occupational Safety and Health (NIOSH) considered Curene 442 (MOCA) a potential human carcinogen (44). Curene 442 has a very low solubility in water. It also has very low vapor pressure. Therefore Curene 442 tends to accumulate in sludges and soils. It is reported to be readily absorbed through skin. Workers handling sludge would, therefore, be at a potential risk from exposure to Curene 442. The Michigan Department of Public Health analyzed urine specimens from Adrian wastewater plant operators for Curene 442 in May, 1979 but found no evidence of its presence. However, Curene 442 was detected in the blood and urine specimens of Anderson Development Company's workers. There are no conclusive epidemiological studies at present on which an evaluation of the carcinogenic risk to Adrian wastewater treatment plant workers can be based.

Several acute toxic effects have been reported by the treatment plant workers at the Morris Forman Plant in Louisville, Ky., and at the North Treatment Plant in Memphis, Tenn., when they were exposed to toxic pesticide chemical wastes. In Louisville, Ky., the exposure was a result of unauthorized use in

March 1977, of a combined sewer for the dumping of a highly toxic pesticide waste material containing hexachlorocyclopentadiene (Hexa) and octachlorocyclopentene (Octa) (38). Workers involved in the steam cleaning of the mechanical screen complained of eye irritation, burning skin above the arms and face, sore throat, and symptoms similar to bronchitis. Airborne concentrations of Hexa at the time of exposure were unknown, but four days after the episode, the air concentrations in the primary treatment areas (screen and grit chambers) ranged from 270 ppb to 970 ppb. By comparison, the recommended TLV for occupational exposure to Hexa is 10 ppb (52). The incident forced the treatment plant to shut down the operation voluntarily from March 29, 1977 to June 5, 1977 resulting in the discharge of untreated sewage containing toxic chemical waste directly into the Ohio River.

In Memphis, Tenn. the situation is not a one-time dumping of toxic chemical waste, but an apparently continuous entry of the wastes into the treatment plant from a pesticide formulator since the treatment plant first opened in 1977. Ironically, the waste material that was dumped into the Louisville sewer system is believed to have originated from the same company. The exposure at both plants is to the same pesticide material, except that in Memphis, Tenn., there are many other chemicals present besides Hexa. Also in Memphis, Tenn., the exposure is one of an almost continuous nature to variable but generally low levels of these chemicals whose long-term toxic effects are not known. Acute effects were reported on several occasions and urinary extraction was demonstrated (10,15).

Eight workers at the wastewater treatment plant in Bloomington, Ind., were exposed to PCBs discharged by the Westinghouse Corporation. Analyses of tissue samples showed that PCBs were present in all eight workers. Two of these workers, who had previously worked at the Westinghouse plant, had PCBs levels of 10.4 ppm and 10.1 ppm (lipid basis). The other six had an average level of 6 ppm. The levels found in these workers are higher than levels found in a national monitoring survey in which only 5% out of a total 637 tissue samples tested had levels higher than 2 ppm. Even though the acute toxicity of PCBs is low, the long-term effects of exposure to PCBs are not known and experiments with laboratory animals have associated PCBs with stillbirths, miscarriages, and various metabolic disorders. The toxic effects of ingestion of PCBs on humans had been demonstrated in a 1968 episode in Japan where a batch of rice oil was accidentally contaminated by PCBs (20,26,45).

There is no information available at present on the health effects of organics on workers involved in the transportation or land spreading, or both, of wastewater or sludge. Information from the study of health effects of pesticides in agricultural use may be useful in this respect. An evaluation of agricultural exposure to pesticides was not included in this review, however.

Effect on Other Populations

No information is available at present on the effects of organics in wastewater and sludge on populations living near wastewater treatment plants. Similarly, no information is available on the effects on populations living near facilities where land application is practiced. At present there are only a few epidemiological studies being conducted on the potential health risks associated with the land

application of wastewater or sludge, or both, to agricultural lands. Among these are: (1) Retrospective and prospective studies in Israel on populations engaged in land application of wastewater (50); (2) a prospective study by Ohio Farm Bureau on potential health effects to farmers involved with land application of sludge (57); and (3) a short-term prospective study of the workers at a wastewater spray irrigation facility in Muskegon, Mich. (11). Of these, only the study at Muskegon, Mich., is taking into consideration the potential health effects of organic chemicals.

Populations in at least two areas have been affected by leachates from industrial waste landfills. In Toone, Tenn., residential drinking water supplies were contaminated by chlorinated pesticide waste chemicals leaching from an old landfill that was used by Velsicol Chemical Corporation for dumping chlorinated pesticide residues from its Memphis, Tenn., operation (39). Some of the symptoms that have been noticed since the fall of 1977, which the residents associate with the contamination of their water supplies, are skin and eye irritation, weakness in joints, and persistent cough (12).

In Love Canal, Niagara Falls, N.Y., where houses and an elementary school were built on an old Hooker Chemical Company landfill site, the chemicals leached into the ground water and were brought back to the surface by rising water tables causing ground-water and air pollution. The rate of miscarriages in this community is reported to be 1 to 5 times higher than expected, and an excess of birth defects was also noticed (46). However, data to support these conclusions have not yet been published. Many of the residents of this community were evacuated from their homes.

Even though the chemical dumps in these two areas are not in use now, they are discharging long-abandoned toxic waste chemicals into air and water. Ironically, Hooker Chemical waste and Velsicol Chemical waste in the past were similar since some of their products were similar.

Water Pollution

Lateral dispersion by runoff can cause surface water pollution by organic chemicals with the possibility of contaminating drinking water supplies. However, there are no published accounts of situations where surface drinking water supplies have been contaminated by organic chemicals in wastewater and sludge used for land application. Surface water pollution by organic chemicals can also result from chemical waste discharges from industries.

Ground-water contamination can occur from collection, treatment, and disposal of municipal wastewater by the following routes: leakage from collecting sewers; leakage from treatment plant lagoons; land disposal of treatment plant effluents and disposal to surface water bodies that recharge aquifers; and land disposal of sludges that are subject to leaching. Although the volume of wastewater entering the ground-water system from these various sources may be substantial, there have been few documented cases of hazardous levels of constituents of sewage affecting well water supplies, largely because the subject has not been studied in detail. A large proportion of the sewage treatment plant effluents discharged to land do not meet secondary treatment standards (19).

The potential for ground-water contamination by long-term land application was investigated in studies sponsored by the United States Environmental

Protection Agency of several land application sites that have been in operation for 10 years or longer. Numerous chemical and biological parameters were monitored in these studies (31). The sites chosen included rapid infiltration systems located at Vineland, N.J. (28), and Milton, Wisc. (3), and slow rate irrigation systems located at Roswell, N.M. (29), and Dickinson, N.D. (4). The wastewater received either primary or secondary treatment prior to the land application. The pesticides endrin, lindane, methoxychlor, toxaphene, 2,4-dichlorophenoxyacetic acid (2,4-D), and 2-(2,4,5-trichlorophenoxy)-propionic acid (2,4,5-TP, Silvex) were monitored in the applied wastewater effluent and in ground water near the land application sites. The concentrations of endrin, methoxychlor, and toxaphene were found to be below the detectable limits in test and control site samples. The herbicides 2,4-D and 2,4-5-TP were found in about the same concentration range as in control samples and were far lower

TABLE 4.—Lindane Concentrations in Applied Effluent and in Ground Water at Land Application Test Sites and at Control Sites,[a] in nanograms per liter

Location (1)	Applied Effluent		Ground Water at Test Site		Ground Water at Control Site		Reference (8)
	Range (2)	Average (3)	Range (4)	Average (5)	Range (6)	Average (7)	
Vineland, N.J.[b,c,d]							
Basin I	120 to 10,300	2,830	—	453	0.4 to 70.4	21.3	28
Basin II	0.5 to 4,510	1,227	—	1,173[e]	0.4 to 70.4	21.3	28
Basin III	0.5 to 4,510	1,227	—	227	0.4 to 70.4	21.3	28
Milton, Wisc.[b,f]	3 to 159	44	0.2 to 1,360	91.7	0.2 to 20.7	7.4	3
Roswell, N.M.[g,h]	49 to 1,250	560	0.1 to 850	74.3	0.1 to 49	10.5	29
Dickinson, N.D.[f,i]	8.3 to 1,690	397	0.4 to 82	9.7	0.4 to 392	53.6	4

[a] National Interim Primary Drinking Water Standard for lindane is 4,000 ng/L.
[b] Land application by rapid infiltration.
[c] Primary treatment followed by infiltration in basins.
[d] Test site samples are taken under the infiltration basins.
[e] Test and control site concentrations are significantly different at the 0.05 level.
[f] Secondary treatment followed by lagoon storage.
[g] Land application by slow rate irrigation.
[h] Secondary treatment followed by chlorination.

than the National Interim Primary Drinking Water Standards proposed by the United States Environmental Protection Agency for these compounds (53). At three of the four sites, lindane was found in the nearby ground water at concentration levels that were significantly higher than the background groundwater concentration, and that at one site was sometimes also above the National Interim Primary Drinking Water Standard (Table 4).

Effect on Food Chain

Only a limited amount of information is available on the effects on the food chain of the organic chemicals present in wastewater and sludge used in land application. Wastewater application methods are important in assessing potential food contamination. Methods of field application of wastewater and sludge can

be one of a variety of surface or subsurface application methods (43). The choice of method depends on the individual land application facility, geographic location, and climate.

Primary attention should probably be given to organic compounds that are known to be potentially toxic and carcinogenic, such as organochlorine pesticides, polycyclic aromatic hydrocarbons, PCBs, etc. The organochlorine compounds are of particular concern because most of them have been found to be known or suspected carcinogens in animal models, are highly resistant to degradation, and are lipid soluble. This accounts for their accumulation and translocation to animals and humans by the food chain.

Physical Contamination.—Chaney and Lloyd (7) have shown that sewage sludges are effectively retained on the top of the vegetation when applied by spraying, and that they are not well removed by rain. Because of the more dilute nature (and lower viscosity) of wastewater and effluents, lower contamination levels on vegetation would be expected. However, evaporation and repeated application may provide sufficient buildup of contaminants to be a cause for concern. Contaminants physically retained on crops may be ingested by animals, or possibly directly by humans if improper practices are followed (5,24).

Uptake by Plants.—The extent to which organic chemicals in wastewater and sludge will be taken up by plants and may be toxic to human beings through food crops is not well understood (5,47). The Michigan Department of Agriculture has analyzed fresh produce grown in household gardens close to Anderson Development Company in Adrian, Mich. (35). As mentioned before, Curene 442 (MOCA), a potential human carcinogen, was found in discharges from the Anderson Development Company and in samples of garden soils in the residential area surrounding the Anderson Development Company. It is believed that these household gardens did not receive municipal sludge from the Adrian wastewater treatment plant as a fertilizer, even though the wastewater treatment plant sludge was given away to home gardeners up until 1977. In a press release dated July 13, 1979, the Michigan Department of Agriculture stated that the results of analysis of limited samples tested in their laboratory showed no evidence of Curene 442 (MOCA) residues in thoroughly washed samples of onion tops and bulbs, zucchini, and radishes.

The uptake of pesticides and PCBs from soils into plants has been studied to a limited extent. Plants do take up many pesticides through roots and translocate them within the plant (40). Some pesticides are taken up, but others are not or are taken up at low levels (41). The pesticides heptachlor, dieldrin, and chlordane are taken up by soybean plants from soil, translocated to the seed, and stored at low levels in the oil (36). In contrast, PCBs are not carried to the top of the soybean plants if direct vapor transmission is prevented by a barrier over the soil (17). Uptake of PCBs by root crops, however, has been demonstrated under field conditions. It was shown that carrot peel contained 97% of the root residue and little was found in the foliage (23). Lawrence and Tosine (30) reported that the PCB concentration in the leaves of corn and grass grown on sludge-treated land was comparable to that in the sludge, whereas the Center for Disease Control (CDC) found negligible amounts of PCBs in vegetation grown in land treated with sludge containing PCB levels as high as 300 ppm in Bloomington, Ind. (26). For example, CDC found that beets grown in sludge-amended soil containing 4 ppm PCBs were found to have 0.6 ppm PCBs

(1/7th of that in the soil), grass from a field containing 8.5 ppm PCBs in soil showed 1.16 ppm PCBs (1/7th of that in the soil), and rhubarb grown in the same soil had 0.25 ppm PCBs (1/34th of that in the soil). The Bloomington sludge was much more heavily contaminated with PCBs than the sludge used by Lawrence and Tosine (30), and was spread on land at 25 tons per acre, which was about three times the maximum application rate recommended (4,21). In order to protect the public health, FDA has recommended to EPA that sludges should not contain more than 10 ppm PCBs on dry weight basis for the application of sludge to agricultural lands (24,58), and EPA adopted this value in its regulation (59). However, municipalities without a significant source of PCBs would be expected to have less than 10 ppm PCBs in their sludges. In a survey of 16 American cities (18), only two cities, Chicago, Ill., and Schenectady, N.Y., had PCBs levels over 10 ppm in sludge. The presence of 300 ppm PCBs in the sludge of Bloomington, Ind., is due to the direct discharge of PCBs waste by Westinghouse Electric Corporation into the city sewerage system, where it is then concentrated in the sludge (26). The levels in Schenectady, N.Y., are high because of the location of two General Electric plants which use large amounts of PCBs in the manufacture of electrical capacitors.

The leaves of brome grass were analyzed for the pesticides methoxychlor, toxaphene, 2,4-D, and 2,4,5-TP at the land application site in Dickinson, N.D. (4). There were no differences in the levels of methoxychlor and toxaphene between control and test sites. Concentrations of 2,4-D and 2,4,5-TP were found to be 15.4 ng/g and 7.0 ng/g at the test sites, whereas the control site samples contained 10.4 ng/g of 2,4-D and 6.6 ng/g of 2,4,5-TP; however there was no mention of whether these differences were statistically significant.

Uptake by Animals.—Cattle and other animals ingest soil as well as plants while grazing. Chemical contaminants, especially metals, pesticides, and PCBs, may build up in selected tissue of such animals when sewage sludge is used on land and pastures (5,47,51). Although wastewater effluent may present a lesser problem, those from highly contaminated sources may result in the buildup of contaminants in the surface layer of soil similar to that observed with sludges.

Some efforts have been made to utilize sewage sludge as an animal feed supplement because of its nutritive value. Sewage sludge subjected to gamma radiation at a dosage of 1 megarad or more to eliminate the risk from pathogens and parasites was used as cattle feed supplement, and the tissue accumulation of heavy metals and pesticide residues was studied by Smith, et al. (51). Pesticide residues were determined in adipose tissue from kidneys. They found that feed consumption was decreased but there was no evidence of toxicity or impaired reproduction. Five of the pesticide residues were significantly elevated by the diet containing 20% sewage sludge. Subsequent feeding on a conventional finishing diet reduced these levels, but not to the levels of the control animals (Table 5).

In the Bloomington, Ind., incident of contamination of wastewater and sludge by PCBs (26), PCBs were found in ground beef from cattle which grazed on contaminated lands. The cattle of a farmer, whose pasture had been flooded by a creek which had been contaminated by PCBs, suffered weight loss and one calf was born without pupils in its eyes. The milk fat of another farmer's cow who grazed on sludge-treated pasture was found to contain 5 ppm PCBs—twice the FDA's 2.5 ppm tolerance level. Now the FDA has lowered the tolerance

level in milk fat to 1.5 ppm, because data developed since 1973 show that PCBs are more toxic than was previously believed (48,49).

Uptake by Humans.—Although there is no direct evidence of human health hazards from uptake through the food chain of the organic chemicals in wastewater and sludge used in land application, some information is available from two incidents of accidental food contamination by PCBs in Japan and PBBs in Michigan.

Food Contamination by PCBs in Japan.—Sporadic outbreaks of a peculiar skin disease characterized by acneiform eruptions and pigmentation of the skin and nails were reported in Japan in 1968. The disease was found to be associated with the consumption of a particular batch of one brand of rice oil. The disease was called Yusho (rice-oil disease). The cause of Yusho was traced to a specific batch of rice oil shipped by one company. The oil was found to contain approx 2,000 ppm of Kanechlor KC400, a commercial preparation of PCB containing 48% chlorine. The latency period between ingestion of the oil and onset of clinical symptoms was generally 5 months to 6 months. Elevated serum triglycerides, increased urinary 17-ketosteroid excretion, and respiratory distress are some of the physiological abnormalities that were observed in Yusho patients (20). Occupational exposure of PCBs had previously been linked to chloracne and hepatic dysfunction.

TABLE 5.—Some Pesticide Residues in Adipose Tissue of Cattle Fed Irradiated Sludge[a]

Treatment group (1)	Residues, in parts per billion of adipose tissue (fresh weight basis)				
	p,p'-DDE (2)	Dieldrin (3)	Oxychlordane (4)	TNC (5)	Arochlor 1254 (6)
Controls	20	20	0	0	0
	20	<20	0	<10	500
	30	<20	0	<10	500
$\bar{x} \pm \sigma$	23 ± 6	<20	0	<10	333 ± 289
Fed diet containing 20% sludge	120	80	70	80	1,380
for 68 days	100	50	50	70	960
	90	50	120	70	980
$\bar{x} \pm \sigma$	103 ± 15	60 ± 17	80 ± 26	73 ± 6	1,107 ± 237
Fed diet containing 20% sludge	60	50	30	20	600
for 68 days followed by conventional diet for additional	20	<20	0	0	500
56 days	60	30	30	20	670
$\bar{x} \pm \sigma$	47 ± 23	33 ± 15	20 ± 17	13 ± 12	590 ± 85
Irradiated dried sludge	40	36	0	0	1,440

[a]Adapted from G. S. Smith, et al. (51).

PBB Episode in Michigan.—The accidental contamination of Michigan dairy farm animal feed by the polybrominated biphenyl (PBB) compound Fire Master BP-6 in 1973–1974 was followed by illness and death of many cattle which

had consumed the PBB-containing feed. Widespread human exposure resulted from consumption of contaminated milk, eggs, meat, and poultry. In response to concerns regarding reports of the appearance of adverse health effects among Michigan dairy farmers exposed to PBBs, and the possible association of such symptoms to their PBBs exposure, Anderson, et al. (1) conducted comprehensive clinical and laboratory examinations of PBB-exposed Michigan farmers. A comparable group of Wisconsin farm families who were not exposed to PBB were used as controls. The symptoms noted were neurological (marked tiredness, fatigue, sleepiness, weakness, poor memory, loss of appetite); musculoskeletal (arthritis-like symptoms); and gastrointestinal (abdominal pain, diarrhea, and weight loss). In most of the participants in the survey, the symptoms represented a distinct change from their previous health pattern. Statistical analysis revealed that the Michigan group has significantly higher prevalence of these symptoms. No clear relationship can be established between serum PBB level and the presence of any of the symptoms noted. The existing difference could not be explained without considering an etiologic role for exposure to PBBs.

These examples are isolated cases of accidental contamination. As such, they represent effects of acute exposure and may not be representative of potential contamination by organic chemicals of food chains from land application. Nevertheless, they serve the purpose of providing some insight into the effect of consuming these chemicals through food chains, and they may be useful in setting standards.

The Curene 442 (MOCA) contamination incident illustrates how an industry can expose the surrounding population to unknown risks (35). The residents of the community surrounding the Anderson Development Company in Adrian, Mich. have been exposed to Curene 442 (MOCA), a potential human carcinogen, due to widespread contamination of roadside soil and air, and household dust. The residents of this community have consumed produce grown in the household gardens. However, laboratory analyses by the Michigan Department of Agriculture showed that Curene 442 (MOCA) was not taken up by the plants that were tested. The Michigan Department of Public Health has monitored the urine specimens of randomly selected persons living within a five block radius of Anderson Development Company, and found no evidence of Curene 442 (MOCA) in urine of the study participants. The long-term health effects of exposure to Curene 442 (MOCA) on the residents of the community of Adrian, Mich., are not known at present.

Control of Organic Chemicals

Control of organic chemicals can be in the form of regulations controlling discharges to wastewater treatment systems, discharge standards for organics in wastewater effluents, or standards for the receiving bodies of water for toxic organic chemicals. The United States EPA has proposed effluent standards for toxic contaminants (60,61) and water quality standards for some organic chemicals that would form the basis for state water quality standards (62-64). These criteria are an indication of the maximum concentration of a pollutant in water which, when not exceeded, will assure protection of specified water use. Dacre proposed SPLVs (soil pollutant limit values) to control the hazard loads of toxic chemicals in soils (13). These values may be compared with

Threshold Limit Values (TLVs), which refer to airborne concentrations of substances, and represent conditions under which it is believed that nearly all workers may be repeatedly exposed day after day without adverse effect. The control measures should also take into account acceptable daily intake (ADI) values estimated by World Health Organization for some pesticide chemicals (65,68).

Conclusions

At present there is insufficient information to assess the long-term health risks associated with exposure to organics in wastewater treatment plants or at land application sites. Acute toxic effects are readily recognizable. The harmful effects thereof can be most directly reduced by enforcement of appropriate pretreatment requirements. If these are not effective, harmful effects can possibly be reduced by removal of workers from the source of toxic fumes, by proper ventilation, and by the use of appropriate respirators and protective clothing, since the exposure to organic chemicals is mainly by inhalation. Effects of exposure to low levels of organics over a long period of time (chronic exposure), including dose-response relationships and health effects, are not known at this time.

Land application systems tend to concentrate chemicals in the soil, enhancing the possibility of uptake by plants and animals. If more complete information on the concentration and fate of the organic chemicals in the environment (water, air, food crops, animals, etc.) were known, it would be possible to make an assessment of the potential hazards associated with the land application of wastewaters and sludges containing toxic organic chemicals. At the present time, sufficient information is not available to do this.

The potential for ground-water pollution from leaching of waste landfills is a serious problem, and adequate preventive measures should be instituted. The potential for air pollution from landfills that have received a variety of substances which may interact with each other has been given little attention.

Further research is needed in the following areas:

1. Uptake of organic chemicals by food crops should be studied in detail. Since little is known about the organics uptake by plants, all parts of the plant should be studied. Some selectivity regarding the parts of the plant can be exercised, depending upon which parts of a given crop are edible.
2. The extent of ground-water pollution by organic chemicals leaching from landfills or sludge applied lands, or both, should be studied.
3. Information should be obtained on pertinent chronic human health effects from agricultural use of and occupational exposure to pesticides. This might be helpful in predicting health effects on treatment plant workers or populations exposed to pesticides in wastewater or sludge, or both.
4. Long-term follow-up should be initiated on populations who have had acute short-term exposure to organic chemicals from waste materials. Examples of such populations are those associated with exposures at Love Canal, N.Y., Toone-Teague, Tenn., and the Louisville, Ky., and Memphis, Tenn. Wastewater Treatment Plants.

5. The potential health effects of utilizing domestic sewage sludge as a cattle feed supplement for both beef and dairy cattle should be studied further.

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